



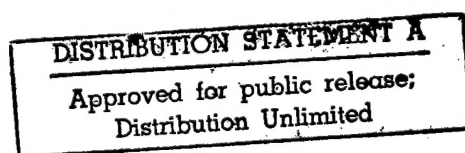
Soil Remediation Demonstration Project: Biodegradation of Heavy Fuel Oils

Charles M. Reynolds, Prasanta Bhunia, and Brent A. Koenen

August 1997

19971030 012

DTIC QUALITY INSPECTED 2



Abstract: Treatment of oil-contaminated soils is necessary to protect water supplies, human health, and environmental quality; but because of limited funds, cleanup costs are often prohibitive. High costs are exacerbated in cold regions such as Alaska, where spills are often in areas inaccessible to heavy equipment and where there is limited infrastructure. Owing to the lack of infrastructure, widespread fuel distribution systems, and the need for heating in the cold climate, there are numerous small-scale oil spills. Low-cost treatments applicable to small-scale spills are needed. The object of this CPAR project was to examine using cost-effective, on-site bioremediation techniques for heavy-oil-contaminated soil in cold regions. Both heavy-oil and

diesel-contaminated soils were used to compare landfarming, a low-intensity treatment, to pile bioventing, a costlier treatment. For each soil-contaminant combination, we compared nutrient additions to a control with no nutrient additions. Under the conditions of this study, landfarming with nutrient additions was as effective for treating diesel-contaminated soil as was bioventing with nutrient additions. For heavy oils, landfarming with nutrients resulted in lower soil concentrations after one year, but differences among treatments were not statistically significant. Because landfarming does not require pumps, electricity, or plumbing, all costs are less than for bioventing. The minimal requirements for infrastructure also make landfarming attractive in remote sites typical of cold regions.

How to get copies of CRREL technical publications:

Department of Defense personnel and contractors may order reports through the Defense Technical Information Center:

DTIC-BR SUITE 0944
8725 JOHN J KINGMAN RD
FT BELVOIR VA 22060-6218
Telephone 1 800 225 3842
E-mail help@dtic.mil
msorders@dtic.mil
WWW <http://www.dtic.dla.mil/>

All others may order reports through the National Technical Information Service:

NTIS
5285 PORT ROYAL RD
SPRINGFIELD VA 22161
Telephone 1 703 487 4650
1 703 487 4639 (TDD for the hearing-impaired)
E-mail orders@ntis.fedworld.gov
WWW <http://www.fedworld.gov/ntis/ntishome.html>

A complete list of all CRREL technical publications is available from

USACRREL (CECRL-LP)
72 LYME RD
HANOVER NH 03755-1290
Telephone 1 603 646 4338
E-mail techpubs@crrel.usace.army.mil

For information on all aspects of the Cold Regions Research and Engineering Laboratory, visit our World Wide Web site:
<http://www.crrel.usace.army.mil>



**US Army Corps
of Engineers®**

Cold Regions Research &
Engineering Laboratory

Soil Remediation Demonstration Project: Biodegradation of Heavy Fuel Oils

Charles M. Reynolds, Prasanta Bhunia, and Brent A. Koenen

August 1997

DTIC QUALITY INSPECTED 3

Prepared for
OFFICE OF THE CHIEF OF ENGINEERS

Approved for public release; distribution is unlimited.

PREFACE

This report was prepared by Dr. Charles M. Reynolds, Research Physical Scientist, Geochemical Sciences Division, Research and Engineering Directorate, U.S. Army Cold Regions Research and Engineering Laboratory, Dr. Prasanta Bhunia, Project Manager, Weston and Sampson Engineers, Inc., and Brent A. Koenen, Environmental Protection Specialist, Geochemical Sciences Division, CRREL.

The objective of the Construction Productivity Advancement Research (CPAR) demonstration project, *Soil Remediation Demonstration Project: Biodegradation of Heavy Fuel Oils*, was to demonstrate, evaluate, and increase the knowledge base for using cost-effective, on-site bioremediation techniques for heavy-oil-contaminated soil in cold and seasonally cold regions. To achieve these objectives, a Project Team was developed that included representatives from the private sector, CRREL, the U.S. Army Engineer District, Alaska, the University of Alaska, and the Alaska Department of Environmental Conservation. The CPAR industry partner, Weston and Sampson Engineers, Inc., worked closely with their cooperator, AGRA Earth and Environmental, Inc., who conducted the site implementation and operation activities. The site was accessible to visitors and tours throughout the demonstration.

The authors acknowledge the contributions of others to this project: Dr. I.K. Iskandar, Chief, Geochemical Sciences Division; D.L. Hardy, U.S. Army Engineer District, Alaska, who provided user perspective at the initial planning meeting; and B. Thomas, formerly with the Alaska Department of Environmental Conservation, who assisted with regulatory requirements.

A significant amount of laboratory and field work was required to accomplish the project goals. The expertise of L.B. Perry and C.S. Pidgeon, who conducted much of the laboratory analysis, was an essential part of this project. R.N. Bailey, J.P. Lariviere, E. Cuthbertson, and J. Velazquez also were essential in conducting the laboratory analyses. B.G. Harrington provided the data-logger support and P. Robinson assisted in the field work. Both MAJ M. Meeks and LTC S. Wagner provided field support and assistance in Alaska. Technical review of this report was provided by Dr. C.J. Martel and S. Hardy, both of CRREL.

The contents of this report are not to be used for advertising or promotional purposes. Citation of brand names does not constitute an official endorsement or approval of the use of such commercial products.

EXECUTIVE SUMMARY

The objective of the Construction Productivity Advancement Research (CPAR) project, *Soil Remediation Demonstration Project: Biodegradation of Heavy Fuel Oils*, was to demonstrate, evaluate, and increase the knowledge base for using cost-effective, on-site bioremediation techniques for heavy-oil-contaminated soil in cold and seasonally cold regions. To accomplish this objective, CRREL and Weston and Sampson Engineers, Inc., conducted a side by side comparison of the commonly used bioventing treatment and the lower-cost landfarming treatment for both diesel- and heavy-oil-contaminated soils.

Treatment of oil-contaminated soils is necessary to protect water supplies, human health, and environmental quality; but, because limited resources are causing increasingly greater competition for funds, cleanup costs are often prohibitive. High costs are exacerbated in cold regions, such as Alaska, where the spills are often in areas inaccessible to heavy equipment and where there is limited infrastructure. Because of the lack of infrastructure, widespread fuel distribution systems, and the need for heating in a cold climate, there are numerous small-scale oil spills in cold regions. Low-cost treatment strategies for these sites are needed.

Our focus was to compare systems applicable to small-scale spills and see if a relatively low-intensity method (landfarming) could compete with more a frequently used but costlier treatment (pile bioventing), without the need for heavy equipment or high energy inputs. Two soils were used to evaluate the two treatment systems: one soil contaminated with heavy oil and the other contaminated with diesel. Diesel-contaminated soils have been shown to respond to biotreatment, both in landfarming and bioventing. Biotreatment of soils contaminated with heavier oils, such as no. 6 heating oil, bunker C fuel oil, and crude oil, have not been evaluated to the same degree as diesel-fuel-contaminated soil. Because a greater proportion of heavy oils is composed of recalcitrant compounds that have limited water solubility, heavy oils are less readily bioremediated than is diesel fuel.

For each soil-contaminant combination, we compared added nutrients to a control with no nutrients. Under the condition of this study, landfarming with nutrient additions was as effective for treating diesel-contaminated soil as was bioventing with nutrient additions. For heavy oils, landfarming with nutrients resulted in lower average soil concentrations after 1 year, but the differences among treatments was not statistically significant. The average Total Petroleum Hydrocarbon (TPH) concentrations for each treatment after 362 days are shown in the following table.

Soil TPH concentrations (mg kg⁻¹) for each treatment after 362 days.

	<i>Heavy Oil</i> [*]		<i>Diesel</i> [†]	
	<i>Landfarming</i>	<i>Bioventing</i>	<i>Landfarming</i>	<i>Bioventing</i>
+Nutrients	4262	4736	1664	1447
-Nutrients	4959	4985	2430	1581

*Initial TPH value, 6157 mg kg⁻¹.

†Initial TPH value, 8348 mg kg⁻¹.

Under the condition of this evaluation, landfarming resulted in significantly greater initial remediation rates for both the heavy-oil and diesel-contaminated soils than did bioventing, and this effect was most pronounced with nutrient additions. In general, diesel-contaminated soil was more effectively treated than the heavy-oil-contaminated soil.

Nutrient additions increased the effects of landfarming for both heavy-oil- and diesel-contaminated soil, but only influenced the diesel-contaminated soil in the bioventing treatments.

For soils contaminated by heavy oils, landfarming resulted in greater bioremediation rates than did the bioventing treatment during the initial treatment stages. Although bioventing was ineffective in reducing the contaminant levels in the initial phase of treatment for the heavy-oil-contaminated soil, bioventing appeared to have stimulated microbial activity and possibly microbial biomass during the initial phase. We believe that increased microbial biomass caused artificially high TPH analyses at the first sampling date, resulting in a zero or negative treatment effect for the first time interval. A similar pattern for crude-oil-contaminated soil was observed during the following warmer season. During the second summer, soil TPH values in the crude-oil-contaminated soil using the bioventing treatment were significantly lower than those for landfarming at the spring sampling time, but appeared to increase by the end of the summer. Continued bioventing may result in lower TPH levels each year of operation, but this cannot be determined by these data.

Because landfarming does not require pumps, electricity, plumbing, or wiring, all costs (capital, maintenance, and operation) are less than for bioventing. The minimal requirements for infrastructure also make landfarming attractive in remote sites typical of cold regions. This project demonstrated that landfarming can compete in both performance and cost for remediation of both heavy-oil- and diesel-contaminated soils at those remote sites.

Soil Remediation Demonstration Project: Biodegradation of Heavy Fuel Oils

CHARLES M. REYNOLDS, PRASANTA BHUNIA, AND BRENT A. KOENEN

INTRODUCTION

Background

There have been numerous petroleum releases in cold, remote regions. Alternatives for cleanup are limited by the remote locations, difficulties in mobilization of heavy equipment, inability to effectively monitor the biotreatment processes, and the costs of site operation. To remediate petroleum-contaminated sites, we need a low-cost, low-input treatment alternative to use in conjunction with existing methods.

In general, indigenous soil microbiota can degrade petroleum compounds. When soils fail to bioremediate at optimum rates, it is often a function of the water solubility of the compound and environmental limitations imposed on the microbes. Major limitations to the microbiota are temperatures that are too high or too low, excess or deficient water, insufficient or excessive nutrients, insufficient carbon in a form that microorganisms can use, poor mixing or distribution of the petroleum in the soil, and, for aerobic microorganisms, lack of oxygen (O_2).

The relative effectiveness of different treatment systems will vary over time. For example, lack of oxygen has been believed to be the primary limitation at depth; thus, air-injection technologies are commonly employed to overcome this. However, it is now well established that subsurface (relatively deep) microbial activity is common, and anaerobic biodegradation of water-soluble petroleum takes place without the need to inject air if alternate electron acceptors, such as oxidized species of iron or nitrogen (such as nitrate), are available. The anaerobic processes are significantly slower than aerobic processes, but they are less

expensive than adding air. The approximate trade-off among air-injection technologies, passive biodegradation, and anaerobic biodegradation is time versus resources (Fig. 1).

Bioremediation treatments are successful when limitations are overcome. The key problem, however, is identifying and implementing the most cost-effective means of doing this at isolated, cold sites. Two important aspects in comparing low-cost to more costlier alternatives are time constraints and monitoring difficulties.

In comparing treatments, it is important to measure effectiveness over time. In applying a bioremediation technology to a particular site, it is also important to consider contaminant transport, leaching, and the location of the problem. The rate of treatment must be compared to the rate of leaching and the distance between the contaminated soil and the area where it might cause harm—the potential receptor. For many sites, transport rates and distance to receptors are such that remediation need not be immediate. If a longer time is acceptable for remediation, more treatment options and lower costs can be considered.

A final difficulty in comparing the effectiveness of treatments is that of obtaining accurate measurements in the field. Limitations to effective bioremediation have been identified primarily through laboratory studies. Measuring rates in the field is much more difficult because we cannot control field conditions effectively. In a field study, where we tried to measure bioremediation rates in a 1-acre (0.4-ha) landfarm that we had treated uniformly, we found that the bioremediation rates varied up to seven-fold (Reynolds 1993b).

Diesel-contaminated soils have been shown to

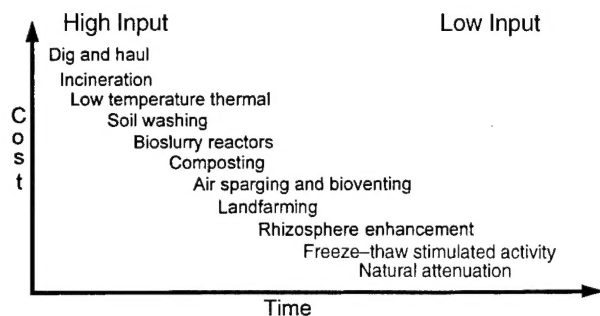


Figure 1. Relative inputs and treatment times for remediation technologies.

respond to biotreatment. Landfarming can be a successful treatment option in cold regions (Reynolds 1993a,b; Reynolds et al. 1994). Bioventing of stockpiled, diesel-contaminated soil has also been used in northern climates, but owing to low temperatures and the distance of many sites from readily accessible highways, logistical, construction, and maintenance requirements and costs are greater than in more temperate climates.

Studies have shown that stimulating soil microbial activity in diesel-contaminated soils in biopiles may raise soil temperatures by 1–3°C (Brar et al. 1993). Also, nutrient amendments to diesel-contaminated soil can enhance bioremediation rates in both landfarming and bioventing (Reynolds 1993b, Brar et al. 1993, Walworth and Reynolds 1995).

Biotreatment of soils contaminated with heavier oils, such as no. 6 heating oil, bunker C fuel oil, and crude oil, have not been evaluated to the same degree as diesel-fuel-contaminated soil. And, because a greater proportion of heavy fuels is composed of recalcitrant compounds that have limited water solubility, these contaminants are less readily bioremediated than diesel fuel.

Objective

The objective of this project was to demonstrate, evaluate, and increase the knowledge base on using cost-effective, on-site bioremediation techniques for treating heavy-oil-contaminated soil in seasonally cold regions.

Approach

The research plan was to compare landfarming and bioventing as treatment technologies for heavy-fuel- and diesel-contaminated soils in cold regions. This was done by conducting side by side comparisons in the field. Because so much less is

known about degradation of heavy-fuel-contaminated soils as compared to diesel-fuel contamination, we expected that a double comparison would provide a better context for analyzing the different techniques.

Project location and soil history

This CPAR project was conducted at the Farmers Loop Permafrost Research Facility in Fairbanks, Alaska. For each biotreatment system (landfarming and ex-situ pile-bioventing), two soils were used. One soil was from a gravel pad area near pump station number 10 on the trans-Alaska pipeline. This soil was contaminated with heavy oil. The other was a sandy soil contaminated with diesel. This material had been used as tank bedding and backfill for a fuel storage tank at Ft. Wainwright, Alaska, and was excavated during tank replacement. These two soils are typical of many contaminated soil sites in northern regions.

FIELD REMEDIATION PROCEDURES

For each contaminated soil, we compared the effects of landfarming to those of bioventing. For all soil-contaminant and treatment scenarios, we also tested the effects of nutrient additions (Table 1).

To comply with regulatory guidelines that were in effect at the start of the field work, treatments were conducted in plots that were lined and bermed. For the landfarming plots, the cells were approximately 3 m square and 60 cm deep. The bioventing plots were approximately 1.5 by 3 m by 100 cm deep. These configurations are representative of both landfarming and bioventing procedures for small-scale spills. Landfarming usually distributes soil in a relatively thin layer that requires a greater area than bioventing, which can use deeper layers of soil.

Table 1. Demonstration factors.

Factor	Levels	Description
Contaminant	2	Heavy-oil, diesel
Treatments	2	Landfarm, bioventing
Subfactors	Levels	Description
Landfarm	2	Control, plus nutrients
Bioventing	2	Control, plus nutrients

Bioventing plots had two evenly spaced air-extraction lines placed horizontally, approximately 45 cm from the surface. The cells were covered by liner material. Air flow was created by pumping a vacuum on the air extraction lines in the bioventing piles. During operation, the pumps were cycled on a schedule of five evenly spaced 1-hour intervals during 24 hours.

Spatial variability

We addressed the problem of spatial variability in soil contamination by using well-mixed contaminated soil in buried nylon "net" bags. To obtain uniform soils for the bags, we removed composite samples from each cell of the two contaminated soils. Approximately 350 subsamples were taken and composited for each soil. Composited samples were homogenized in a large rotary mixer. Because the total amount of the composited sample of each soil was too large to mix at once, it was divided into thirds and each third was mixed for 30 minutes, then the three portions were inter-mixed for 1 hour. The net bags permitted mass and energy fluxes throughout the soil both inside and outside of the bags, yet allowed us to recover the soil for time-series sampling. For each treatment, we used enough bags for triplicate samples at each time-series sampling.

For the landfarm, each bag was cylindrical and measured approximately 5 cm in diameter and 40 to 50 cm in length. We made the net cylinders by rolling a portion of netting around a hollow metal form until two complete layers of netting covered the form, and then tying a knot in the netting to secure the bottom. The net-wrapped form was filled with the well-mixed contaminated soil. After filling, the hollow form was removed from the netting and the sample was placed back into the field soil.

This procedure let us obtain relatively uniform field data for this study. It provided a uniform initial contaminant concentration level in each bag with a sufficient number of bags in each cell for replicate, time-series sampling.

Nutrient amendments

Nutrients were uniformly spread on the surface of appropriate cells. Commercially available agricultural fertilizer, granular 20-20-10, was used. The nitrogen form was ammonium (NH_4^+). We applied fertilizer to the surface because this method, with a minimum of labor and machinery, is the most likely to be used in the field. Application rates were approximately 0.62 kg m^{-2} (0.13 lb ft^{-2}) on

the landfarm and approximately 1.23 kg m^{-2} (0.25 lb ft^{-2}) on the bioventing cells. Twice as much fertilizer was applied to the bioventing cells because they were nearly twice as deep as the landfarming cells. This resulted in similar amounts of nutrients added on a mass:mass basis.

Sampling and analysis

After 54 days, triplicate sample bags were randomly removed from each cell and sampled for Total Petroleum Hydrocarbon (TPH) fractions. TPH extractions from the soil were done by sonication with methylene chloride. Anhydrous sodium sulfate was added to the soil during extraction as a drying agent. Extracts initially were analyzed by gravimetric analysis and then by resolution in methylene chloride followed by gas chromatography and flame ionization detection (GC-FID). For TPH, chromatograms were integrated from baseline to baseline.

Costs

Estimates for this project were based on treating 500 yd^3 (382 m^3) for 1 year and ranged from $\$5\text{--}\11 yd^{-3} ($\$3.82\text{--}\8.41 m^{-3}) for landfarming and $\$20\text{--}\23 yd^{-3} ($\$15.30\text{--}\17.59 m^{-3}) for bioventing. Cost breakdowns are provided in Table 2.

In this demonstration, all soil was excavated from other sites and transported to the demonstration site at Farmers Loop; therefore, soil handling costs were the same for both treatments. Other costs that were equal for the two treatments include acquisition and application of nutrients and, for this demonstration, monitoring.

Costs that were incurred in bioventing, but not landfarming, included plumbing for air flow, blowers, wiring, electricity for operation, periodic monitoring of the blowers, and maintenance of the blowers. In practice, in-situ landfarming may be applicable at many sites because heavier oils and more recalcitrant compounds are typically less mobile. In-situ treatment would remove both liner and soil handling costs.

Remediation costs are site specific and absolute values derived from a particular site can be transferred to other sites only with caution. Typically, costs are reported per ton or per cubic yard, but consideration must be given to the fixed costs, such as mobilization and decontamination of equipment, that are required regardless of the volume treated. With larger volumes of soil treated, fixed costs are distributed over a greater volume of soil and result in a lower per unit cost.

Table 2. Itemized costs (\$) of treatment options for 500 yd³ (382 m³) of contaminated soil.

Item	Bioventing (percent time blower on)			Landfarming (percent lined and covered)	
	100%	50%	20%	100%	0
Earthwork subcontract	3950	3950	3950	1950	1950
Mobilization/decontamination	200	200	200	200	200
Electrical subcontract	2760	2760	2760	N/A	N/A
System piping (venting, water)	750	750	750	0	0
Bottom liner	480	480	480	2400	0
Regenerative blower	1000	1000	1000	N/A	N/A
Polyethylene cover	250	250	250	250	0
Electrical power (\$0.11 per kW)	1445	723	301	0	0
Electrical start-up draw (15% of total electrical)	217	108	45	0	0
Nutrients	450	450	450	450	450
Total	11,502	10,671	10,186	5250	2600
Total cost per yd ³	23	21	20	11	5
Total cost per m ³	17.58	16.06	15.29	8.41	3.82

RESULTS AND DISCUSSION

Initial samples and contaminant homogeneity

Measuring field bioremediation rates is difficult because of the nonuniformity of contaminant distribution; it is also costly. Yet, bioremediation rate comparisons, based on direct field measurements, are the true measure of treatment effects. To effectively compare landfarming to bioventing, we must either develop a better system for field monitoring, or wait a longer time before measuring so that differences would be more pronounced, or know that the effects of the treatments would be vastly different. Because the effect of the treatments is our unknown and a comparison of rates over time is important, the only acceptable option was to develop a better system for field monitoring.

To determine a bioremediation rate at any given time, we must be able to compare contaminant concentrations to the initial concentration of the contaminant in the soil. To make valid rate comparisons, it is essential either to provide known contaminant concentrations at the start (t_0) or take an extremely large number of samples to quantify the random error caused by spatial variability. Because of the "destructive nature" of taking samples (i.e., you cannot actually resample the same volume of soil) and the difficulty in determining exactly how many samples would be required to sufficiently reduce the variability, we chose to use a method that provided uniform sample concentrations at the start, as described in the *Sampling and Analysis* section.

Random samples taken at the beginning of the field study were analyzed for TPH components to determine the variability at t_0 . Results (Table 3) show that distribution of TPH and other petroleum fractions had a Coefficient of Variation (CV) less than 5%, were homogeneous, and demonstrated that using mixed soil in permeable bags provides uniform t_0 samples. Thus, this technique provided equal starting concentrations on which to base remediation rates.

On the basis of previous studies of relative rates of degradation, the diesel soil was expected to degrade faster than the heavy oil because of the smaller percentage of more recalcitrant, heavy-oil compounds.

Initial incubation period: elapsed time 0 to 54 days, 29 July to 21 September 1995

Diesel

For diesel-contaminated soil, all treatments decreased relative to the t_0 values during the first 54 days of treatment (Fig. 2). Landfarming with nutrients was the most effective and had significantly ($P < 0.05$) lower TPH concentrations at t_{54} than did the other treatments. TPH concentrations in the landfarming without nutrient treatment also decreased relative to the t_0 values and had lower ($P < 0.10$) TPH concentrations than the bioventing treatments, but not to the same degree as TPH concentrations when nutrients were added. Bioventing treatments, both with and without nutrients, were less effective than the landfarm-

Table 3. Concentrations (mg kg⁻¹) at t_0 (methylene chloride extraction + sonication).

Fraction	Heavy-oil mean		Diesel mean	
TPH _{gravimetric}	11389	±309	8480	±267
TPH _{gas chromatography}	6072	±266	8348	±291

ing treatments during the t_0 to t_{54} interval. Because diesel has a larger percentage of volatile and mobile constituents than heavy oil, the t_0 to t_{54} data suggested that volatilization and diffusion accounted for a portion of the initial losses.

Heavy oil

During the t_0 to t_{54} interval, decreases in TPH concentrations were much less in the heavy-oil-contaminated soil than in the diesel-contaminated soil (Fig. 3). We observed the same ranking for efficacy of treatments as we did for the diesel-contaminated soil; landfarming with nutrient amendments again was statistically the best treatment. In contrast to the diesel-contaminated soil, there was essentially no decrease in heavy oil TPH levels in the bioventing treatments during the initial treatment phase.

Cold season incubation: elapsed time, 54 to 238 days, 21 September 1995 to 16 May 1996

Diesel

TPH concentrations decreased for all treatments during t_{54} to t_{238} (Fig. 2). During a significant portion of this time, the soil was frozen. However, an unusual warm period in September 1995 kept the soil warmer for longer than normal and this probably contributed to the decrease in TPH levels during this period. The rates of TPH decrease during this time were not significantly different for the two landfarming treatments and averaged 13.6 mg kg⁻¹ day⁻¹. TPH reduction rates for the two bioventing treatments were also similar to each other, averaging 23.8 mg kg⁻¹ day⁻¹. However, the TPH reduction rates in bioventing treatments were significantly greater than in landfarming treatments during this time. From our data, we cannot sepa-

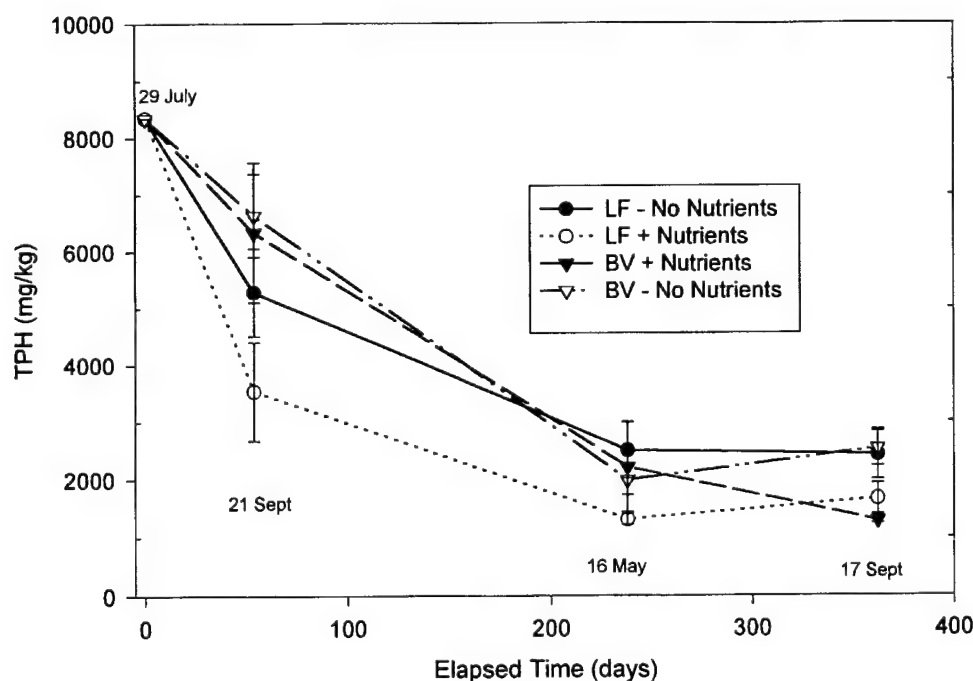


Figure 2. TPH reduction, diesel (LF = landfarming; BV = bioventing).

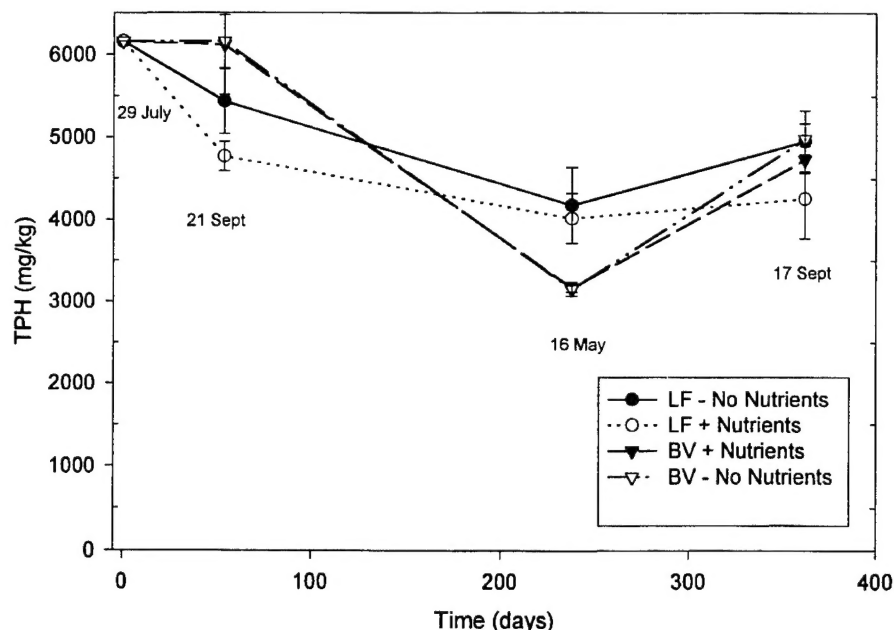


Figure 3. TPH reduction, heavy oil (LF = landfarming; BV = bioventing).

rate TPH reductions that may have been caused by biodegradation during the early portion of this time, or transport processes that may have been influenced by freezing and freeze-thaw cycles during the winter and early spring. Some samples taken on 16 May 1996 were still partially frozen, suggesting that temperatures sufficiently high for significant biodegradation to occur following soil thawing had not yet been attained. The TPH concentrations for the diesel-contaminated soil on 16 May were not significantly different ($P < 0.05$) for any of the treatments.

Heavy oil

TPH concentrations decreased more rapidly for both of the bioventing treatments than for either of the landfarming treatments during the cold-season phase of the operation (Fig. 2). Similar to the diesel-contaminated soil, the nutrient-amended and no-nutrient bioventing treatments decreased TPH concentrations in the heavy-oil-contaminated soil at similar rates, averaging $16.2 \text{ mg kg}^{-1} \text{ day}^{-1}$. TPH reduction rates in the landfarming treatments were only $5.5 \text{ mg kg}^{-1} \text{ day}^{-1}$ during this time. In contrast to the diesel-contaminated soil, the samples taken on 16 May from the bioventing treatments in the heavy-oil-contaminated soil had significantly lower TPH concentrations than did the landfarming treatments. Because these samples were from a greater depth than the landfarm-

ing samples, the lag-time preceding soil freezing may have allowed longer treatment time in the bioventing cells vs. the landfarming cells.

Summer incubation: elapsed time, 238 to 362 days, 16 May to 17 September 1996

Diesel

Nutrient amendments appeared to have a greater effect than either landfarming or bioventing on the TPH concentrations by t_{362} (Fig. 2). TPH concentrations in the nutrient-amended bioventing treatment continued to decrease during the summer and at t_{362} were less than those in the nutrient-amended landfarming treatment. There was little change from t_{238} to t_{362} in the treatments where no nutrients had been applied.

Heavy oil

Changes in the TPH concentrations in the heavy oil concentrations from t_{238} to t_{362} are noticeable; the TPH concentrations appear to actually increase in both bioventing treatments and remain constant in the landfarming treatments (Fig. 3). Landfarming with nutrients tended towards greater TPH reduction than the other three treatments, but this effect was not significant. The variability in TPH concentrations at t_{362} for both landfarming treatments was greater than for either bioventing treatments, suggesting that the processes causing

TPH reduction were more responsive to a variable that was not well controlled.

DISCUSSION

After 362 days of treatment, landfarming was as effective in reducing TPH concentrations in diesel-contaminated soil as was bioventing. Nutrient additions had a significant effect on reducing diesel concentrations for both landfarming and bioventing.

For heavy-oil-contaminated soil, the effect of nutrient additions was less evident and there were no significant differences among any treatments at t_{362} . For both bioventing treatments, measured TPH concentrations increased from t_{238} to t_{362} . Several factors may account for this. When using TPH as an indicator of treatment effects, many have observed an apparent increase in TPH concentrations at the onset of biotreatment. We hypothesize that initial increases in measured TPH are caused by an increase in microbial biomass that accompanies a growing microbial population and precedes measurable decreases in TPH concentrations. During the initial phases of degradation, the microorganisms use bioavailable forms of native soil carbon rather than contaminant carbon. Native soil carbon forms are less likely to be included in TPH extractions or measurements. However, the metabolites produced by the rapidly expanding population do appear as part of the TPH measurement. As the microbial population increases, measured TPH concentrations increase as bioavailable native soil carbon becomes depleted with no decrease in contaminant carbon in the soil. TPH concentrations would not decrease until after bioavailable carbon has been consumed and contaminant carbon is used. Subsequent mineralization of the contaminant carbon would then eventually result in a decrease in soil contaminant TPH concentrations.

CONCLUSIONS

This demonstration showed that landfarming can be a reasonable alternative to bioventing for both heavy-oil- and diesel-contaminated soils at remote sites typical of cold regions. In diesel-contaminated soil, reduction in TPH concentrations was greatest using landfarming with nutrient amendments. Landfarming with nutrient additions should be considered as a treatment op-

tion at sites where it can be easily used.

Annual reductions in TPH concentrations for the heavy oil after approximately 1 year were not significantly different for any of the treatments. For heavy-oil-contaminated soil, landfarming was more effective than bioventing during the initial phase of treatment, and this effect was more pronounced when nutrients were added. The lag period that occurs prior to rapid microbial growth and increased activity appeared to have a greater effect in the heavy-oil-contaminated soil compared to the diesel-contaminated soil. We hypothesize that the measured increase in TPH concentrations is an analytical artifact of microbial fatty acids and related microbial biomass that are produced during adaptation of the soil microbiota to new conditions and carbon sources. If this is the cause, it is reasonable to predict that, given favorable incubation conditions that are sufficiently long, there may be a period of rapid heavy-oil bioremediation following the temporary increase in measured TPH values. However, if the favorable incubation time is too short owing to a brief summer season, the same cycle may occur in subsequent years. Low-cost strategies that further stimulate the microbial population may be needed to effectively treat these soils. Recent studies suggest that plant-based systems may be useful (Nichols et al. 1997).

These data demonstrate that, depending on the nature of the contaminant and nutrient status of the soil, the brevity of the summer season—when temperatures are favorable for microbial activity—may prevent either unamended or nutrient-amended biotreatment from attaining sufficient microbial numbers and activity to substantially reduce TPH concentrations during that season. This may result from microbial population cycling, from low populations at early summer to high populations at late summer, with little TPH metabolized. The persistence of the heavier fraction of older oil spills in cold regions suggests that this may be happening (Collins et al. 1993). For diesel spills, the effect is not as pronounced and many of the compounds are more readily degraded; hence, nutrient amendments appeared to provide sufficient stimulation to significantly reduce TPH concentrations.

Despite both treatments being done *ex situ* (requiring excavation and transportation of soil), costs for the landfarming treatment were less than for bioventing. The reduced requirements for infrastructure also make landfarming attractive in remote sites typical of cold regions. Landfarm installation and operation in a remote area is less

costly than bioventing. Similarly, the remoteness of many contaminated sites may allow more time for the bioremediation process, which would favor a low-input, low-maintenance landfarming system.

Substantial cost savings may be realized in using landfarming at many remote sites, but absolute dollar savings can not be predicted without knowing the specifics for each site. The costs for application of any soil treatment technology are site dependent, and the cost savings derived from using landfarming vary with both site specifics and the alternative cleanup strategies that may be available.

RECOMMENDATIONS

Because of lower costs and treatment that was as effective as bioventing, landfarming should be given consideration for treating heavy-oil-contaminated soils in cold regions. Land-farming has lower capital, operation, and maintenance costs than bioventing and may be used in situ on some surface-contained spills. Surface applied nutrients can be used and mixing by tilling is not essential. Additionally, landfarming is a robust system that can recover from harsh environmental changes, such as freezing and drying. This demonstration has shown that it is effective for both diesel and heavy-oil contamination.

COMMERCIALIZATION PLAN

The products of this project include data that demonstrate landfarming and bioventing to be equally successful in treating heavy-oil-contaminated soil. Additionally, there have been visitors at the site to see the landfarming and bioventing processes. Weston and Sampson Engineering and Consulting and AGRA Earth and Environmental now have experience plus data and cost figures to present to potential clients. Regional, national, and international conferences where presentations covering this demonstration has been made or submitted are listed below.

- *Cold Regions Remediation Conference, BP Exploration (Alaska)*, 26–27 March 1997, Anchorage, Alaska.
- *Second Tri-Service Environmental Technology Workshop*, 10–12 June 1997, St. Louis, Missouri.
- *U.S. Army Corps of Engineers, Innovative Tech-*

nologies HTRW Workshop, 17–21 March 1997, Las Vegas, Nevada.

- *Fifth International Symposium on Cold Region Development*, American Society of Civil Engineers—Technical Council of Cold Regions Engineering, 4–10 May 1997, Anchorage, Alaska.

- *Fourth International Symposium, In-Situ and Onsite Bioreclamation*, 28 April–1 May 1997, New Orleans, Louisiana.

- *European Union Workshop on Soil Remediation*, 16–19 November 1996, Rothamsted, UK.

- *Third Annual International Petroleum Environmental Conference*, 24–27 September 1996, Albuquerque, New Mexico.

- *Soil Science Society of America*, 3–8 November 1996, Indianapolis, Indiana.

LITERATURE CITED

Brar, G.S., P.M. Currier, and C.M. Reynolds (1993) Remediation of petroleum contaminated soils with bioventing in cold regions. *Superfluid XIV Conference*, 29 November–1 December, Washington, D.C.

Collins, C.M., C.H. Racine, and M.E. Walsh (1993) Fate and effects of crude oil spilled on subarctic permafrost terrain in interior Alaska: Fifteen years later. USA Cold Regions Research and Engineering Laboratory, CRREL Report 93-13.

Nichols, T.D., D.C. Wolf, H.B. Rogers, C.A. Beyrouy, and C.M. Reynolds (1997) Rhizosphere microbial populations in contaminated soils. *Water, Air, and Soil Pollution*, 95: 165–178.

Reynolds, C.M. (1993a) Contaminated soil treatment in cold regions. *American Public Works Association Reporter*, July, p. 26.

Reynolds, C.M. (1993b) Field measured bioremediation rates in a cold region landfarm: Spatial variability relationships. In *Proceedings of the Seventh Annual East Coast Conference on Hydrocarbon Contaminated Soils* (P.T. Kostecki and E.J. Calabrese, Eds.) Chelsea, Michigan: Lewis Publishers, p. 487–499.

Reynolds, C.M., M. Travis, W.A. Braley, and R.J. Scholze (1994) Applying field expedient bioreactors and landfarming in cold climates. In *Second International Symposium, In-Situ and Onsite Bioreclamation*. Chelsea, Michigan: Lewis Publishers, p. 100–106.

Walworth, J.L., and C.M. Reynolds (1995) Bioremediation of a petroleum contaminated soil: Effects of phosphorus, nitrogen, and temperature. *Journal of Soil Contamination*, 4(3): 299–310.

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestion for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE August 1997		3. REPORT TYPE AND DATES COVERED	
4. TITLE AND SUBTITLE Soil Remediation Demonstration Project: Biodegradation of Heavy Fuel Oils				5. FUNDING NUMBERS	
6. AUTHORS Charles M. Reynolds, Prasanta Bhunia, and Brent A. Koenen					
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) U.S. Army Cold Regions Research and Engineering Laboratory 72 Lyme Road Hanover, New Hampshire 03755-1290				8. PERFORMING ORGANIZATION REPORT NUMBER Special Report 97-20	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Office of the Chief of Engineers Washington, D.C. 20314-1000				10. SPONSORING/MONITORING AGENCY REPORT NUMBER	
11. SUPPLEMENTARY NOTES For conversion of SI units to non-SI units of measurement consult ASTM Standard E380-93, <i>Standard Practice for Use of the International System of Units</i> , published by the American Society for Testing and Materials, 1916 Race St., Philadelphia, Pa. 19103.					
12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution is unlimited. Available from NTIS, Springfield, Virginia 22161				12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) Treatment of oil-contaminated soils is necessary to protect water supplies, human health, and environmental quality; but because of limited funds, cleanup costs are often prohibitive. High costs are exacerbated in cold regions such as Alaska, where spills are often in areas inaccessible to heavy equipment and where there is limited infrastructure. Owing to the lack of infrastructure, widespread fuel distribution systems, and the need for heating in the cold climate, there are numerous small-scale oil spills. Low-cost treatments applicable to small-scale spills are needed. The object of this CPAR project was to examine using cost-effective, on-site bioremediation techniques for heavy-oil-contaminated soil in cold regions. Both heavy-oil and diesel-contaminated soils were used to compare landfarming, a low-intensity treatment, to pile bioventing, a costlier treatment. For each soil-contaminant combination, we compared nutrient additions to a control with no nutrient additions. Under the conditions of this study, landfarming with nutrient additions was as effective for treating diesel-contaminated soil as was bioventing with nutrient additions. For heavy oils, landfarming with nutrients resulted in lower soil concentrations after one year, but differences among treatments were not statistically significant. Because landfarming does not require pumps, electricity, or plumbing, all costs are less than for bioventing. The minimal requirements for infrastructure also make landfarming attractive in remote sites typical of cold regions.					
14. SUBJECT TERMS Biotreatment Bioventing Crude oil Diesel Land farming Petroleum				15. NUMBER OF PAGES 16	
				16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL		